

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

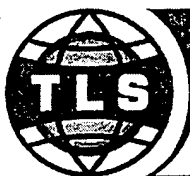
Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**



# **Technical Language Service**

Translations From And Into Any Language

## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 2003 - 154539 A**

**Polyester Resin Composition Used For Calendering,  
and a Sheet Using the Same**

**Your Ref: 102203 - 14**

**For: Eastman Chemical Company**

---

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-973-6009 . email: sales@tls-translations.com

(19) Japanese Patent Office (JP)

(11) Unexamined Patent Application No:

(12) Unexamined Patent Gazette (A)

**Kokai 2003-154539**

**(P2003-154539A)**

(43) Date of Publication: May 27, 2003

(51) Int. Cl. <sup>7</sup>	Class. Symbols	FI	Subject Codes (Reference)
B 29 C 43/24		B 29 C 43/24	4F071
C 08 J 5/18	CFD	C 08 J 5/18	4F204
C 08 K 5/00		C 08 K 5/00	4J002
C 08 L 67/02		C 08 L 67/02	
// B 29 K 67:00		B 29 K 67:00	

Request for Examination: Not yet submitted

Number of Claims: 10 OL

Total of 11 pages [in original]

(Continued on last page)

(21) Application No.: 2001-326741  
(P2001-326741)

(22) Date of Filing: October 24, 2001

(31) Priority Application No.: 2001-274010  
(P2001-274010)

(32) Priority Date: September 10, 2001

(33) Priority Country: Japan (JP)

(71) Applicant: 000003160  
Toyobo Co., Ltd.  
2-8 Doshimahama 2-chome, Kita-ku, Osaka-shi, Osaka-fu

(72) Inventor: Kazunori Komatsu  
c/o Toyobo General Research Institute  
1-1 Katata 2-chome, Otsu-shi, Shiga-ken

(72) Inventor: Osamu Iritani  
c/o Toyobo General Research Institute  
1-1 Katata 2-chome, Otsu-shi, Shiga-ken

(72) Inventor: Mitsuo Nishida  
c/o Toyobo General Research Institute  
1-1 Katata 2-chome, Otsu-shi, Shiga-ken

(Continued on last page)

(54) [Title of the Invention]

**Polyester Resin Composition Used For  
Calendering, and a Sheet Using the Same**

(57) [Summary]

[Object] To provide a polyester resin composition having excellent release properties from the rolls during fabrication, and that enables calendering of a high-quality sheet in which the finished sheet has good transparency and excellent solvent resistance and detergent resistance.

**[Means of Achievement]** A polyester resin composition used for calendering, comprising at least two types of polyester and a lubricant, wherein the polyester resin composition used for calendering is characterized in that at least one of the two types of polyester consists of a non-crystalline polyester.

**[Claims]**

**[Claim 1]** A polyester resin composition used for calendering, comprising at least two types of polyester and a lubricant, wherein the polyester resin composition used for calendering is characterized in that at least one of the two types of polyester consists of a non-crystalline polyester.

**[Claim 2]** The polyester resin composition used for calendering according to claim 1, characterized in comprising 50 to 97 weight parts of a non-crystalline polyester, 3 to 50 weight parts of a crystalline polyester, and 0.01 to 5 weight parts of a lubricant.

**[Claim 3]** The polyester resin composition used for calendering according to claims 1 or 2, characterized in that the non-crystalline polyester has an aromatic dicarboxylic acid with a carbon number of 8 to 14 and an aliphatic or alicyclic glycol with a carbon number of 2 to 10 as the main components thereof.

**[Claim 4]** The polyester resin composition used for calendering according to claim 3, characterized in that the aromatic dicarboxylic acid with a carbon number of 8 to 14 in the non-crystalline polyester is terephthalic acid or isophthalic acid.

**[Claim 5]** The polyester resin composition used for calendering according to claims 3 or 4, characterized in that the aliphatic or alicyclic glycol with a carbon number of 2 to 10 in the non-crystalline polyester is ethylene glycol, diethylene glycol, neopentyl glycol, or cyclohexanedimethanol.

**[Claim 6]** The polyester resin composition used for calendering according to any of claims 1 through 5, characterized in that the melting point of the crystalline polyester is 90 to 220°C.

**[Claim 7]** The polyester resin composition used for calendering according to any of claims 1 through 6, characterized in that the glass transition temperature of the crystalline polyester is -100 to 30°C.

**[Claim 8]** The polyester resin composition used for calendering according to any of claims 1 through 7, characterized in comprising 50 mol% or more of at least one type from among ethylene glycol, 1,3-propanediol, 1,4-butanediol, 1,6-hexanediol, and cyclohexanedimethanol

when the total quantity of the glycol components in the crystalline polyester is taken to be 100 mol%.

**[Claim 9]** The polyester resin composition used for calendering according to any of claims 1 through 8, characterized in that a metal salt of an organic phosphoric acid ester is used as a lubricant.

**[Claim 10]** A sheet obtained by means of calendering the resin composition according to any of claims 1 through 9.

### **[Detailed Description of the Invention]**

#### **[0001]**

**[Technological Field of the Invention]** The present invention relates to a polyester resin composition that is optimal for calendering; that has excellent release properties from a roll during polyester calendering, long-run workability, and thermal stability; and that is useful in the shrink labeling used for food products, cosmetics, and beverages, as well as in aroma-retaining heat sealed film, oil-resistant multilayer sheeting, and other various sheets and films, and is particularly useful in sheets and films used in industrial processes.

#### **[0002]**

**[Prior Art]** Vinyl chloride sheeting (film) is widely used in conventional practice in a variety of applications because of its low cost and excellent transparency. Extrusion molding and calender molding are known as methods for working this sheeting, but friction with the die lips in extrusion molding affects the workability (moldability) of the resin more than in calender molding, which lowers the precision of the sheet (film) thickness, width, flow direction, and the like, and this method is ill-suited for large-scale production. The latter method is therefore more widely employed when productivity and quality are emphasized. Because the flexibility of vinyl chloride can be freely adjusted by means of the addition of plasticizers, it is easily workable in the form of a sheet, and an example of an application thereof is in decorative sheeting (film) imprinted with wood grain for enhancing the design of wood. However, due to drawbacks whereby dioxins are released during incineration of vinyl chloride, and restrictions on the use of plasticizers due to the presence of endocrine-disrupting chemicals, a changeover from vinyl chloride sheeting (film) to other materials has been under way in recent years. Among substitute

materials, polyester is favored for its physical aspects, cost, and other attributes. However, using polyester as a substitute has two major drawbacks.

[0003] The first drawback is the difficulty of applying polyester to calender molding, which is widely used as a molding method for polyvinyl chloride. Extrusion molding has mainly been used conventionally as a method for manufacturing sheets and films from polyester resin for its ease of fabrication. However, molding is difficult when calendering is attempted using polyester resin, because of its relatively strong adhesion to the rolls when thermoplasticized, and its tendency to adhere to the rolls during fabrication. The addition of various lubricants has therefore been attempted in order to prevent adhesion to the rolls. Lubricants that have thus been investigated include, for example, polyethylene wax, paraffin wax, and other hydrocarbon lubricants, as well as higher fatty acid lubricants, higher alcohol lubricants, metallic soaps made from higher fatty acids, fatty acid amide lubricants, ester lubricants, and various other lubricants, but adequate roll release properties and long-run time have been impossible to obtain, and drawbacks occur during long-run fabrication, whereby the sheet (film) thus obtained is discolored, and the hue of the sheet (film) is adversely affected.

[0004] The second drawback is that the resistance to chemical agents such as solvents and detergents is weak, and whitening and swelling occur when the polyester is dipped and scrubbed therein. The sheet often becomes soiled when wood to which the decorative sheet is affixed is made into tables, furniture, sink retaining doors, and the like. Solvents such as alcohol and methyl ethyl ketone are used to wipe the sheets in the process of removing these stains, but this causes the polyester itself to crystallize and whiten, or if crystallization does not occur, the sheet swells, which ruins its outward appearance and renders it unusable. Removal of stains using solvents is also routinely performed in households and kitchens when such furniture is used in households in general, and the use of solvents to remove stains harms the outward appearance of the sheet in these cases as well.

[0005] Examples will be given below of polyester resin calendering that has hitherto been investigated, and the prior art will be described in further detail. Sheets were obtained by means of calendering a crystalline polyethylene terephthalate elastomer in JP (Kokai) Nos. 7-278418, and 8-283547, but the resin crystallized and the sheets whitened immediately after fabrication, which made them difficult to use in cosmetic applications that require transparency. Sheets were also made by means of calendering a non-crystalline polyethylene terephthalate copolymerized

with cyclohexanedimethanol in JP (Kokai) No. 11-343353, 2000-136294, 2000-186191, 2000-302951, 2001-64496, 2001-4019, and US Patent No. 6068910, and although the sheets had good transparency, all had inadequate resistance to chemicals, the sheets whitened when dipped in solvents, and they were difficult to use in decorative sheeting. The inventors and others have also investigated various admixtures according to these disclosures using this polyester, but the transparency of the finished sheet declines if the quantity of lubricant is increased in order to improve roll release properties, and the proper balance between the quantity of polyester and lubricant has been impossible to achieve. When using these lubricants as main components, drawbacks also occurred whereby the sheet gradually became discolored during long-run fabrication, and the outward appearance of the sheet was adversely affected.

**[0006]**

**[Problems to Be Solved by the Invention]** In view of the foregoing drawbacks, and object of the present invention is to provide a polyester resin composition used for calendering whereby roll release properties during fabrication and aspects of the sheet after fabrication (virtually no coloration, good transparency and printability, and excellent solvent resistance and detergent resistance) can be obtained at the same time.

**[0007]**

**[Means Used to Solve the Above-Mentioned Problems]** As a result of concentrated investigation aimed at achieving the foregoing objects, the inventors discovered that roll release properties during fabrication and aspects of the sheet after fabrication (virtually no coloration, good transparency and printability, and excellent solvent resistance and detergent resistance) can be obtained at the same time by means of admixing a non-crystalline polyester resin, a crystalline polyester resin, and a lubricant, and thus developed the present invention.

**[0008]** Specifically, the present invention has the following characteristics.

(1) A polyester resin composition used for calendering, comprising at least two types of polyester and a lubricant, wherein the polyester resin composition used for calendering is characterized in that at least one of the two types of polyester consists of a non-crystalline polyester.



(2) The polyester resin composition used for calendering according to (1), characterized in comprising 50 to 97 weight parts of a non-crystalline polyester, 3 to 50 weight parts of a crystalline polyester, and 0.01 to 5 weight parts of a lubricant.

[0009] (3) The polyester resin composition used for calendering according to (2), characterized in that the non-crystalline polyester has an aromatic dicarboxylic acid with a carbon number of 8 to 14 and an aliphatic or alicyclic glycol with a carbon number of 2 to 10 as the main components thereof.

[0010] (4) The polyester resin composition used for calendering according to (3), characterized in that the aromatic dicarboxylic acid with a carbon number of 8 to 14 in the non-crystalline polyester is terephthalic acid or isophthalic acid.

[0011] (5) The polyester resin composition used for calendering according to (3) or (4), characterized in that the aliphatic or alicyclic glycol with a carbon number of 2 to 10 in the non-crystalline polyester is ethylene glycol, diethylene glycol, neopentyl glycol, or cyclohexanedimethanol.

[0012] (6) The polyester resin composition used for calendering according to any of (1) through (5), characterized in that the melting point of the crystalline polyester is 90 to 220°C.

[0013] (7) The polyester resin composition used for calendering according to any of (1) through (6), characterized in that the glass transition temperature of the crystalline polyester is -100 to 30°C.

[0014] (8) The polyester resin composition used for calendering according to any of (1) through (7), characterized in comprising 50 mol% or more of at least one type from among ethylene glycol, 1,3-propanediol, 1,4-butanediol, 1,6-hexanediol, and cyclohexanedimethanol when the total quantity of the glycol components in the crystalline polyester is taken to be 100 mol%.

[0015] (9) The polyester resin composition used for calendering according to any of (1) through (8), characterized in that a metal salt of an organic phosphoric acid ester is used as a lubricant.

[0016] (10) A sheet obtained by means of calendering the resin composition according to any of (1) through (9).

[0017]

**[Embodiments of the Invention]** The polyester resin composition of the present invention used for calendering consists of a polyester resin composition used for calendering comprising at least two types of polyester and a lubricant, wherein the polyester resin composition used for

calendering is characterized in that at least one of the two types of polyester consists of a non-crystalline polyester. By comprising at least two types of polyester, roll release properties during fabrication and aspects of the sheet after fabrication (virtually no coloration, good transparency and printability, and excellent solvent resistance and detergent resistance) can be obtained at the same time.

[0018] Furthermore, a product comprising 50 to 97 weight parts of a non-crystalline polyester, 3 to 50 weight parts of a crystalline polyester, and 0.01 to 5 weight parts of a lubricant is preferred. "Non-crystalline" as mentioned herein is defined as follows. Specifically, using a differential scanning calorimeter (DSC), the temperature of the product is raised from  $-100^{\circ}\text{C}$  to  $300^{\circ}\text{C}$  at a rate of  $20^{\circ}\text{C}/\text{minute}$ , the product is cooled from  $300^{\circ}\text{C}$  to  $-100^{\circ}\text{C}$  at a rate of  $50^{\circ}\text{C}/\text{min}$ , and then the temperature is raised again from  $-100^{\circ}\text{C}$  to  $300^{\circ}\text{C}$  at a rate of  $20^{\circ}\text{C}/\text{min}$ . The product is non-crystalline if it does not have a distinct fusion peak during either of the two temperature increases. Alternatively, the product is crystalline if it has a distinct fusion peak as measured by means of DSC under these same conditions.

[0019] The minimum admixed quantity of the non-crystalline polyester is 50 weight parts, preferably 60 weight parts, and more preferably 65 weight parts; and the admixed maximum thereof is 97 weight parts, preferably 95 weight parts. The minimum admixed quantity of the crystalline polyester is 3 weight parts, preferably 5 weight parts; and the admixed maximum thereof is 50 weight parts, preferably 45 weight parts, and more preferably 40 weight parts.

[0020] An admixed quantity of the non-crystalline polyester of less than 50 weight parts, or an admixed quantity of the crystalline polyester of over 50 weight parts is not preferred, because crystallization in the finished sheet becomes severe, which gives it a whitened appearance. Also, if the admixed quantity of the non-crystalline polyester is over 95 weight parts, or if the admixed quantity of the crystalline polyester is less than 3 weight parts, the solvent resistance and detergent resistance of the finished sheet decline. In other words, the crystalline polyester present in the finished sheet must be crystallized to a degree that does not harm the transparency of the sheet itself in order to obtain the solvent resistance that is an object of the present invention. It is predicted that due to the presence of this crystalline component, swelling of the resin does not occur, molecular motion is inhibited, and whitening is prevented even when the surface of the sheet comes in contact with a solvent. To achieve solvent resistance, it is important that at least a

slight fusion peak occur when the finished sheet is measured by means of DSC as described above.

**[0021]** The non-crystalline polyester resin used in the present invention preferably has an aromatic dicarboxylic acid with a carbon number of 8 to 14 and an aliphatic or alicyclic glycol with a carbon number of 2 to 10 as the main components thereof. A "main component" herein is considered to comprise 50 mol% or more, preferably 60 mol% or more, and more preferably 65 mol% or more of each of the dicarboxylic acid component and the glycol component when the total quantity of both of these components is taken to be 100 mol%. The elongation and mechanical properties of the sheet obtained by means of calendering decline if both components comprise less than 50 mol%.

**[0022]** The aromatic dicarboxylic acid with a carbon number of 8 to 14 in the non-crystalline polyester resin used in the present invention is preferably terephthalic acid or isophthalic acid. The elongation and mechanical properties of the sheet obtained by means of calendering are further enhanced when these dicarboxylic acids are used.

**[0023]** Polycarboxylic acids other than the abovementioned terephthalic acid and isophthalic acid may be copolymerized in the non-crystalline polyester resin of the present invention, and examples thereof include orthophthalic acid, naphthalene dicarboxylic acid, succinic acid, adipic acid, azelaic acid, sebacic acid, decane acids, dimer acids, cyclohexane dicarboxylic acid, trimellitic acid, and other known products.

**[0024]** The aliphatic or alicyclic glycol with a carbon number of 2 to 10 in the non-crystalline polyester used in the present invention preferably consists of ethylene glycol, diethylene glycol, neopentyl glycol, or cyclohexanedimethanol. Furthermore, a non-crystalline polyester is preferred that comprises less than 60 mol%, more preferably less than 50 mol%, or particularly preferably less than 40 mol% of at least one type selected from ethylene glycol, neopentyl glycol, and cyclohexanedimethanol. Also, 10 mol% or more, and particularly 15 mol% or more, of at least one type selected from diethylene glycol, neopentyl glycol, and cyclohexanedimethanol are preferably comprised therein. Among these, a combination of ethylene glycol and neopentyl glycol has good compatibility with the lubricant, which makes it easy to obtain both roll release properties and sheet transparency.

**[0025]** The non-crystalline polyester resin of the present invention may be copolymerized with other polyhydric alcohol components other than the abovementioned ethylene glycol, diethylene

glycol, neopentyl glycol, and cyclohexanedimethanol, and examples thereof include 1,2-butanediol, 1,3-butanediol, 1,5-pentanediol, 3-methyl-1,5-pentanediol, 2-methyl-1,3-propanediol, hexanediol, nonanediol, dimer diols, ethylene oxide adducts and propylene oxide adducts of bisphenol A, polyethylene glycol, polypropylene glycol, polytetramethylene glycol, 2-butyl-2-ethyl-1,3-propanediol, tricyclodecane dimethanol, hydroxypivalic acid neopentyl ester, 2,2,4-trimethyl-1,5-pentanediol, trimethylolpropane, and the like.

[0026] The melting point of the crystalline polyester resin used in the present invention is preferably 90 to 220°C. A more preferred minimum is 100°C, more preferably 110°C. At the same time, a preferred maximum is 200°C, more preferably 190°C. If the melting point is less than 90°C, solvent resistance and detergent resistance decline due to reduced crystallinity. On the other hand, if the melting point is over 220°C, the need arises to set the surface temperature of the rolls during calendering to a high level, causing the roll release properties during fabrication to become poor and the sheet elongation to decline due to a reduction in the molecular weight of the polyester by means of hydrolysis.

[0027] The glass transition temperature of the crystalline polyester resin used in the present invention is preferably -100 to 30°C. A more preferred minimum is -80°C, and an even more preferred minimum is -70°C. A more preferred maximum is 25°C. A glass transition temperature of less than -100°C is not preferred, because of strong tackiness in the finished sheet, which leads to blocking between sheets. On the other hand, if a crystalline polyester resin with a glass transition temperature of over 30°C is used, the sheet rapidly cools to about room temperature during the cooling process following the calendering process, and solvent resistance may decline due to inadequate progress in the crystallization of the crystalline polyester resin.

[0028] The crystalline polyester used in the present invention preferably comprises 50 mol% or more of at least one type from among ethylene glycol, 1,3-propanediol, 1,4-butanediol, 1,6-hexanediol, 1,6-hexanediol, and cyclohexanedimethanol when the total quantity of the glycol components in the crystalline polyester is taken to be 100 mol%. 55 mol% or greater is more preferred, and 60 mol% or more is even more preferred. The crystalline polyester present in the sheet must be promptly crystallized in order to demonstrate solvent resistance, but the above-mentioned components preferably comprise 50 mol% or more to ensure adequate crystallinity. If these components comprise less than 50 mol%, solvent resistance declines. Among the above-mentioned components, 1,3-propanediol and 1,4-butanediol are particularly preferred from the

perspective of solvent resistance, because crystallization is rapidly accelerated when these components are used.

[0029] The crystalline polyester resin used in the present invention may be copolymerized with other polycarboxylic acid components and polyhydric alcohol components other than the above-mentioned ethylene glycol, 1,3-propanediol, 1,4-butanediol, 1,6-hexanediol, and cyclohexane dimethanol, and examples of polycarboxylic acid components include terephthalic acid, isophthalic acid, orthophthalic acid, naphthalene dicarboxylic acid, succinic acid, adipic acid, azelaic acid, sebacic acid, decanoic acid, dimer acids, cyclohexane dicarboxylic acid, trimellitic acid, and other known products. On the other hand, examples of polyhydric alcohol components include 1,2-butanediol, 1,3-butanediol, 1,5-pentanediol, 3-methyl-1,5-pentanediol, 2-methyl-1,3-propanediol, nonanediol, dimer diols, ethylene oxide adducts and propylene oxide adducts of bisphenol A, polyethylene glycol, polypropylene glycol, polytetramethylene glycol, 2-butyl-2-ethyl-1,3-propanediol, tricyclodecane dimethanol, hydroxypivalic acid neopentyl ester, 2,2,4-trimethyl-1,5-pentanediol, trimethylolpropane, and the like.

[0030] The number-average molecular weight of the non-crystalline polyester resin and crystalline polyester resin used in the present invention is preferably 15,000 to 40,000, more preferably 18,000 to 35,000, and even more preferably 20,000 to 35,000. If the number-average molecular weight is less than 15,000, the strength and elongation of the sheet is inadequate, due to inadequate resin cohesion, which makes it brittle and unusable. On the other hand, because the melt viscosity rises if the number-average molecular weight is 40,000 or above, the optimal temperature for calendering also rises, which results in poor roll release properties in the sheet.

[0031] The acid value of the non-crystalline polyester resin and crystalline polyester resin used in the present invention is preferably  $60 \text{ eq}/10^6 \text{ g}$  or lower, more preferably  $50 \text{ eq}/10^6 \text{ g}$  or lower, and even more preferably  $40 \text{ eq}/10^6 \text{ g}$  or lower. If the acid value is more than  $60 \text{ eq}/10^6 \text{ g}$ , hydrolysis is further accelerated when the resin is heated during calendering, and the mechanical strength of the finished sheet is reduced. The roll release properties of the sheet also decline as decomposition of the resin progresses.

[0032] The melt viscosity of the polyester resin composition used in the present invention is preferably 6,000 to 60,000 dPa·sec when the temperature is  $220^\circ\text{C}$  and the shear speed is  $100 \text{ sec}^{-1}$ , more preferably 7,000 to 50,000 dPa·sec, and even more preferably 8,000 to 40,000 dPa·sec. If the melt viscosity is less than 6,000 dPa·sec, the roll release properties of the

sheet decline due to increased adhesion of the resin. On the other hand, a melt viscosity of over 60,000 dPa·sec is too high, and the resultant decrease in productivity makes it impractical. When a sheet is made by means of calendering from the polyester resin of the present invention, a lubricant must be admixed with the polyester resin in order to improve the roll release properties of the molten sheet.

[0033] The admixed quantity of the lubricant used in the present invention is preferably 0.01 to 5 weight parts. A more preferred minimum is 0.05 weight parts, an even more preferred minimum is 0.1 weight parts, and the most preferred minimum is 0.5 weight parts. Also, a more preferred maximum is 4.5 weight parts, an even more preferred maximum is 4 weight parts, and the most preferred maximum is 3.5 weight parts. If the quantity of the lubricant is less than 0.01 weight parts, enhancement to the roll release properties is difficult to obtain, and if the quantity is more than 5 weight parts, the transparency, coloration, and printability of the processed sheet show a tendency to decline

[0034] Lubricants used in the present invention include polyolefinic waxes, metal salts of organic phosphate esters, organic phosphate esters, ester compounds of adipic acid or azelaic acid with higher aliphatic alcohols; ethylene bis-stearamides, methylene bis-stearamides, ethylene bis-oleamides, and other fatty acid amides; ester compounds of glycerin with higher aliphatic acids, ester compounds of pentaerythritol with higher aliphatic acids, higher aliphatic alcohols, higher aliphatic acids, paraffins derived from petroleum or coal, waxes, natural or synthetic polyester waxes, metallic soaps made from higher aliphatic acids, and the like. These may be used in combinations of one, two, or more types thereof. Using a polyolefinic wax and/or metal salt of an organic phosphate ester is particularly preferred, because the roll release properties of the sheet and the transparency of the finished sheet can be easily obtained at the same time.

[0035] Polyolefinic waxes used as lubricants in the present invention include polyethylene wax, polypropylene wax, and derivatives of these, and these derivatives include copolymers with other monomers, such as acrylic acid, vinyl acetate, styrene, and maleic acid, and partially oxidized decomposition products.

[0036] Metal salts of organic phosphate esters used as lubricants in the present invention include, for example, metal salts of the organic phosphate ester indicated by the general formula (I) below and/or metal salts of the organic phosphate ester indicated by the general formula (II) below.

**[0037]** Formula (I):  $[\{RO(C_fH_{2f}O)_n\}_{3-a-o}PO(O)_a(OH)_e]_d\{M(OH)_b\}_c$

(In the formula, R is a hydrocarbon group with a carbon number of 4 to 30; M is an alkali metal, alkaline earth metal, Zn, or Al; a is 1 or 2; e is 0 or 1 (0 or 1 when a is 1, and 0 when a is 2); b is 0 to 2; c is 1 or 2; d is 1 to 3; f is 2 or 3; n is 0 to 60; and a, b, c, and d have the following relation with the valence (hereinafter referred to as m) of the metal (M). When  $m = 1$ , then  $b = 0$ ,  $d = 1$ , and  $a = c$ ; when  $m = 2$ , then  $b = 0$ ,  $c = 1$ , and  $a \times d = 2$ , or  $b = 1$ ,  $d = 1$ , and  $a = c$ ; when  $m = 3$ , then  $b = 0$ ,  $d = 3$ ,  $a = c$ ,  $b = 1$ ,  $c = 1$  and  $a \times d = 2$ , or  $b = 2$ ,  $d = 1$ , and  $a = c$ ; furthermore, when  $m \geq 2$ , mutually different phosphate ion groups may be bonded with the metal (M), and in this case,  $d = 2$  or  $3$  is the total number of mutually different phosphate ion groups. Also, when  $d$  is 2 or 3, each of the structures enclosed in brackets may be mutually the same or different.)

**[0038]** Formula (II):  $[\{R^1O(C_fH_{2f}O)_n\}_{3-a-e}PO(O)_a(OH)_e]_d\{M(OCOR^2)_s(OH)_x\}_t$

(In the formula,  $R^1$  is a hydrocarbon group with a carbon number of 4 to 30;  $R^2$  is an alkyl group with a carbon number of 1 to 25; M is an alkali metal, alkaline earth metal, Zn, or Al; a is 1 or 2; e is 0 or 1 (0 or 1 when a is 1, and 0 when a is 2); d is 1 or 2; s is 1 or 2; x is 0 or 1; t is 1 or 2; f is 2 or 3; and n is 0 to 60. Also,  $s + x = 1$  or  $2$ ; and a, d, s, and t have the following relation with the valence (hereinafter referred to as m) of the metal (M). When  $m = 2$ , then  $s = 1$ ,  $d = 1$ , and  $a = t$ ; when  $m = 3$ , then  $s = 1$ ,  $t = 1$ , and  $a \times d = 2$ , or  $s = 2$ ,  $d = 1$ , and  $a = t$ ; and furthermore, when  $m = 3$ , mutually different phosphate ion groups may be bonded with the metal (M), and in this case,  $d = 2$  is the total number of various phosphate ion groups. Also, when  $d$  is 2, each of the structures enclosed in brackets may be mutually the same or different.)

**[0039]** An alkyl group, phenyl group, arylalkyl group, alkenyl<sup>1</sup> group, or alkylphenyl group is preferred for the hydrocarbon group with a carbon number of 4 to 30 indicated by R in the abovementioned general formula (I) and the hydrocarbon group with a carbon number of 4 to 30 indicated by  $R^1$  in the abovementioned general formula (II). Also, Li, Na, K, and the like, for example, are preferred for the alkali metal indicated by M in the general formulae (I) and (II), and Mg, Ca, Ba, and the like, for example, are preferred as alkaline earth metals.

**[0040]** The metal salt of the organic phosphate ester indicated by the general formula (I) and the metal salt of the organic phosphate ester indicated by the general formula (II) may be manufactured by means of the usual method, and no particular limit is placed on the manufacturing method.

**[0041]** Preferred examples of metal salts of the organic phosphate ester indicated by the general formula (I) include chemical compounds (1) through (13) in Table 1 below, chemical compounds (14) through (16) in Table 2 below, and the like, and preferred examples of metal salts of the organic phosphate ester indicated by the general formula (II) include chemical compounds (15) through (26) in Table 2 below. These may be used singly or together in combinations of two or more types thereof.

**[0042]** Also, as shown by decimal fractions included in the number of repetitions of oxyethylene or oxytrimethylene units in the polyethyl alcohol component, these compounds (compounds (1) through (26)) are pure compounds or mixtures of a plurality of metal salts of phosphate esters having different numbers of repeating oxyethylene units or oxytrimethylene units in the polyether alcohol component (the number of repetitions (n) in  $(C_1H_2O)_n$  in the formula).

---

<sup>1</sup> Translator's note: The original has "argenyl," which appears to be a typographical error.



[0043]

[Table 1]

Compound No.	Structural Formula	Compound No.	Structural Formula
(1)	$\text{C}_{16}\text{H}_{33}(\text{OCH}_2\text{CH}_2)_{13}\text{O}-\text{P}(=\text{O})(\text{OH})\text{ONa}$	(9)	$[\text{C}_{20}\text{H}_{41}(\text{OCH}_2\text{CH}_2)_{10.5}\text{O}]_2-\text{P}(=\text{O})(\text{OH})\text{OZnOH}$
(2)	$\text{C}_{17}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{20.3}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$	(10)	$[\text{C}_{20}\text{H}_{41}(\text{OCH}_2\text{CH}_2)_{4.0}\text{O}]_2-\text{P}(=\text{O})(\text{OH})\text{OZnOH}$
(3)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{18.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$		$\text{C}_{20}\text{H}_{41}(\text{OCH}_2\text{CH}_2)_{4.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(4)	$\text{C}_{17}\text{H}_{37}(\text{OCH}_2\text{CH}_2)_{6.6}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OMgOH}$		$[\text{C}_{10}\text{H}_{21}(\text{OCH}_2\text{CH}_2)_{10.5}\text{O}]_2-\text{P}(=\text{O})(\text{OH})\text{OMgOH}$
(5)	$\text{C}_{14}\text{H}_{25}(\text{OCH}_2\text{CH}_2)_{4.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$	(11)	$\text{C}_{11}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{4.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(6)	$[\text{C}_{12}\text{H}_{25}(\text{OCH}_2\text{CH}_2)_{7.1}\text{O}]_2-\text{P}(=\text{O})(\text{OH})\text{OK}$		$\text{C}_{18}\text{H}_{35}(\text{OCH}_2\text{CH}_2)_{5.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OLi}$
(7)	$[\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{2.8}\text{O}]_2-\text{P}(=\text{O})(\text{OH})\text{Zn}$		$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{6.6}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OZnOH}$
(8)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{6.0}\text{O}-\text{P}(=\text{O})(\text{OH})\text{Zn}$	(13)	$\text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_{6.6}\text{O}-\text{P}(=\text{O})(\text{OH})\text{OZnOH}$

[0044]

[Table 2]

Compound No.	Structural Formula	Compound No.	Structural Formula
(14)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2\text{CH}_2)_5.4\text{O}-\text{P}(=\text{O})(\text{OH})\text{ONa}$	(21)	$\text{C}_{12}\text{H}_{25}(\text{OCH}_2\text{CH}_2)_6.3\text{O}-\text{P}(=\text{O})(\text{O})_2-\text{Al}(\text{O})_2\text{COO}-\text{C}_7\text{H}_{15}$
(15)	$\left[ \left( \text{C}_9\text{H}_{19} \right)_2 \text{C}_6\text{H}_4 (\text{OCH}_2\text{CH}_2)_5.2\text{O}-\text{P}(=\text{O})(\text{O})_2\text{O}-\text{C}_a \right]_2$	(22)	$\left[ \text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_7.0\text{O}-\text{P}(=\text{O})(\text{O})_2\text{O}-\text{CH}_3\text{COO}-\text{AlOH} \right]_2$
(16)	$\text{C}_{24}\text{H}_{49}\text{O}-\text{P}(=\text{O})(\text{O})_2-\text{Zn}$	(23)	$\left[ \text{C}_{18}\text{H}_{33}(\text{OCH}_2\text{CH}_2)_8.3\text{O}-\text{P}(=\text{O})(\text{O})_2\text{O}-\text{Zn} \right]_2$
(17)	$\text{C}_{14}\text{H}_{29}(\text{OCH}_2\text{CH}_2)_7.8\text{O}-\text{P}(=\text{O})(\text{OH})\text{O}-\text{Ba}-\text{O}$	(24)	$\text{C}_6\text{H}_5(\text{OCH}_2\text{CH}_2)_{18.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{O}-\text{Ca}-\text{O}$
(18)	$\text{C}_9\text{H}_{19}(\text{OCH}_2\text{CH}_2)_5.0\text{O}-\text{P}(=\text{O})(\text{OH})\text{O}-\text{Mg}-\text{O}$	(25)	$\text{C}_{18}\text{H}_{37}(\text{OCH}_2\text{CH}_2\text{CH}_2)_{11.2}\text{O}-\text{P}(=\text{O})(\text{OH})\text{O}-\text{Ba}-\text{O}$
(19)	$\text{C}_8\text{H}_{17}(\text{OCH}_2\text{CH}_2)_{18.5}\text{O}-\text{P}(=\text{O})(\text{OH})\text{O}-\text{Ca}-\text{O}$	(26)	$\left( \text{C}_{15}\text{H}_{31}\text{O} \right)_2\text{P}(=\text{O})(\text{O})_2-\text{Zn}-\text{O}$
(20)	$\left[ \text{C}_{13}\text{H}_{27}(\text{OCH}_2\text{CH}_2)_7.0\text{O}-\text{P}(=\text{O})(\text{O})_2\text{O}-\text{Zn} \right]_2$		

[0045] The polyester resin of the present invention is preferably used in the form of a composition in which an antioxidant is admixed therein in order to minimize thermal degradation of the polyester resin (prevent coloration of the resin, adhesion to the rolls, and other events due to thermal degradation) during fabrication. Phenol-type antioxidants, organic phosphite ester compounds, and the like, for example, are suitable antioxidants.

[0046] Specific examples of the phenol-type antioxidant used in the present invention include 2,6-di-tert-butylphenol, 2,6-di-tert-butyl-4-methylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,4,6-tri-tert-butylphenol, 2-tert-butyl-4-methoxyphenol, 3-methyl-4-isopropylphenol, 2,6-di-tert-butyl-4-hydroxymethylphenol, 2,2-bis(4-hydroxyphenyl)propane, bis(5-tert-butyl-4-hydroxy-2-methylphenyl)sulfide, 2,5-di-tert-amylhydroquinone, 2,5-di-tert-butylhydroquinone, 1,1-bis(3-tert-butyl-4-hydroxy-5-methylphenyl)butane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl)methane, 2,6-bis(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methylphenol, bis(3-tert-butyl-4-hydroxy-5-methylbenzyl)sulfide, bis(3-tert-butyl-5-ethyl-2-hydroxyphenyl)methane, bis(3,5-di-tert-butyl-4-hydroxydiphenyl)methane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl)sulfide, 1,1-bis(4-hydroxyphenyl)cyclohexane, ethylene bis[3,3-bis(3-tert-butyl-4-hydroxyphenyl)butyrate], bis[2-(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methyl-6-tert-butylphenyl]terephthalate, 1,1-bis(2-hydroxy-3,5-dimethylphenyl)-2-methylpropane, 4-methoxyphenol, cyclohexylphenol, p-phenylphenol, catechol, hydroquinone, 4-tert-butylpyrocatechol, ethyl gallate, propyl gallate, octyl gallate, lauryl gallate, cetyl gallate,  $\beta$ -naphthol, 2,4,5-trihydroxybutyrophenone, 1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)benzene, 1,6-bis[2-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxy]hexane, tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxymethyl]methane, bis(3-cyclohexyl-2-hydroxy-5-methylphenyl)methane, bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxyethyl]sulfide, n-octadecyl<sup>2</sup>-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate, bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionylamino]hexane, 2,6-bis(3-tert-butyl-2-hydroxy-5-methylphenyl)-4-methylphenol, bis[S-(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)]thioterephthalate, 1,1,3-tris(3-tert-butyl-4-hydroxy-6-methylphenyl)butane, and the like. Also, one, two, or more types of these compounds may be used together.

[0047] The admixed quantity of the phenol-type antioxidant is preferably a maximum of 1.0 weight parts or less, and particularly preferably 0.8 weight parts or less; and a preferred

minimum is 0.01 weight parts or more, and particularly preferably 0.02 weight parts or more. If the admixed quantity is less than 0.01 weight parts, thermal degradation during fabrication is difficult to minimize in an efficient manner, and if the admixed quantity is more than 1.0 weight parts, the effects that minimize thermal degradation are excessive, which is uneconomical.

[0048] Specific examples of organic phosphite ester compounds used in the present invention include triphenyl phosphite, tris(methylphenyl) phosphite, triisooctyl phosphite, tridecyl phosphite, tris(2-ethylhexyl) phosphite, tris(nonylphenyl) phosphite, tris(octylphenyl) phosphite, tris[decylpoly(oxyethylene)] phosphite, tris(cyclohexylphenyl) phosphite, tricyclohexyl phosphite, tri(decyl)thio phosphite, triisodecyl thiophosphite, phenyl-bis(2-ethylhexyl) phosphite, phenyl-diisodecyl phosphite, tetradecyl poly(oxyethylene)-bis(ethylphenyl) phosphite, phenyl-dicyclohexyl phosphite, phenyl-diisooctyl phosphite, phenyl-di(tridecyl) phosphite, diphenyl-cyclohexyl phosphite, diphenyl-isooctyl phosphite, diphenyl-2-ethylhexyl phosphite, diphenyl-isodecyl phosphite, diphenyl-cyclohexyl phenyl phosphite, diphenyl-(tridecyl)thio phosphite, nonylphenyl-ditridecyl phosphite, phenyl-p-tert-butylphenyl-dodecyl phosphite, diisopropyl phosphite, bis[o[c]tadecyl poly(oxyethylene)] phosphite, octylpoly(oxypropylene)-tridecyl poly(oxypropylene) phosphite, monoisopropyl phosphite, diisodecyl phosphite, diisooctyl phosphite, monoisooctyl phosphite, didodecyl phosphite, monododecyl phosphite, dicyclohexyl phosphite, monocyclohexyl phosphite, monododecyl poly(oxyethylene) phosphite, bis(cyclohexylphenyl) phosphite, monocyclohexyl-phenyl phosphite, bis(p-tert-butylphenyl) phosphite, tetratridecyl-4,4'-isopropylidene diphenyl diphosphite, tetratridecyl-4,4'-butylidene bis(2-tert-butyl-5-methylphenyl) diphosphite, tetraisooctyl-4,4'-thiobis(2-tert-butyl-5-methylphenyl) diphosphite, tetrakis(nonylphenyl)-poly(propyleneoxy)isopropyl diphosphite, tetratridecyl-propylene oxypropyl diphosphite, tetratridecyl-4,4'-isopropylidenedicyclohexyl diphosphite, pentakis(nonylphenyl)-bis[poly(propyleneoxy)isopropyl] triphosphite, heptakis(nonylphenyl)-tetrakis[poly(propyleneoxy)isopropyl] pentaphosphite, heptakis(nonylphenyl)-tetrakis(4,4'-isopropylidenediphenyl) pentaphosphite, decakis(nonylphenyl)-heptakis(propyleneoxyisopropyl) octaphosphite, decaphenyl-heptakis(propyleneoxyisopropyl) octaphosphite, bis(butoxycarbomethyl)-2,2-dimethylene-trimethylene dithiophosphite, bis(isooctoxycarbomethyl)-2,2-dimethylene trimethylene dithiophosphite, tetradecyl-ethylene dithiophosphite, tetradodecyl-hexamethylene dithiophosphite, tetradodecyl-2,2'-oxydiethylene

---

<sup>2</sup> Translator's note: The original reads, "otatadecyl," which appears to be a typographical error.

dithiophosphite, pentadodecyl-di(hexamethylene) trithiophosphite, diphenyl phosphite, 4,4'-isopropylidene-dicyclohexyl phosphite, 4,4'-isopropylidenediphenyl-alkyl(C12 to C15) phosphite, 2-tert-butyl-4-[1-(3-tert-butyl-4-hydroxyphenyl)isopropyl]phenyl di(p-nonylphenyl) phosphite, ditridecyl-4,4'-butylidene bis(3-methyl-6-tert-butylphenyl) phosphite, dioctanodecyl 2,2-dimethylene trimethylene diphosphite, tris(cyclohexylphenyl) phosphite, hexatridecyl 4,4',4''-1,1,3-butanetriyl-tris(2-tert-butyl-5-methylphenyl) triphosphite, tridodecyl thiophosphite, decaphenyl-heptakis(propyleneoxyisopropyl) octaphosphite<sup>3</sup>, dibutyl-pentakis(2,2-dimethylene trimethylene) diphosphite, dioctyl-pentakis(2,2-dimethylene trimethylene) diphosphite, didecyl 2,2-dimethylene trimethylene diphosphite, and metal salts thereof with lithium, sodium, potassium, magnesium, calcium, barium, zinc, and aluminum. Also, one, two, or more types of these compounds may be used together.

[0049] The admixed quantity of the organic phosphite ester compound is preferably a maximum of 3.0 weight parts or less, and particularly preferably 2.0 weight parts or less; and a preferred minimum is 0.01 weight parts or more, and particularly preferably 0.02 weight parts or more. If the admixed quantity is less than 0.01 weight parts, thermal degradation during fabrication is difficult to minimize in an efficient manner, and if the admixed quantity is more than 3.0 weight parts, the effects that minimize thermal degradation are excessive, which is uneconomical.

[0050] When a sheet is made by means of calendering from the polyester resin of the present invention, a lubricant must be admixed with the polyester resin in order to improve the roll release properties of the molten sheet.

[0051] It is sometimes preferable to add a crystal nucleator as needed to the polyester resin composition of the present invention. This is because a crystal nucleator increases the rate of crystallization of the crystalline polyester, rapidly brings crystallization to completion, and is capable of controlling the size of spherulites by means of adjusting the number of crystal nuclei. Specifically, sheet transparency and solvent resistance are easily obtained at the same time by means of these adjustments. Specific examples of crystal nucleators include talc, silica, graphite, carbon powder, pyrophyllite, gypsum, neutral clay, and other inorganic particles; magnesium oxide, aluminum oxide, titanium oxide, and other metal oxides; sulfate, phosphate, silicate, oxalate, stearate, benzoate, salicylate, tartrate, and sulfonate salts; salts of montanic acid wax;

<sup>3</sup> Translator's note: The original reads, "octabosphite," which appears to be a typographical error.

salts of montanic acid ester wax; terephthalate and carboxylate salts; and ionic copolymers composed of  $\alpha$ -olefins and  $\alpha,\beta$ -unsaturated carboxylic acids.

[0052] In the polyester resin composition of the present invention, other components may be arbitrarily added according to the application. These components include, for example, fillers, UV absorbers, light stabilizers, pigments, antistatic agents, antimicrobial agents, epoxy<sup>4</sup> compounds, cross-linking agents, sulfur-type antioxidants and the like.

[0053]

[Working Examples] The following working examples will be given in order to describe the present invention in further detail, but the present invention is in no way limited by the working examples. Measured values given in the synthesis examples were found by means of the measurement methods below.

[0054] Resin composition: The product was dissolved in heavy chloroform, and assayed by means of H-NMR.

[0055] Glass transition temperature: Measured using a differential scanning calorimeter by placing a 10-mg measurement sample on an aluminum pan, pressing the lid closed, and measuring at a temperature increase rate of 20°C/min.

[0056] Number-average molecular weight: Found as a polystyrene equivalent value by means of gel permeation chromatography using hexafluoroisopropanol as a solvent.

[0057] Acid value: 1 g of the resin was dissolved in 30 mL of chloroform, and the acid value was found by means of titration with a 0.1 N potassium hydroxide ethanol solution. Phenolphthalein was used as an indicator.

[0058] <Synthesis example of non-crystalline polyester (A)>

960 weight parts of dimethyl terephthalate, 527 weight parts of ethylene glycol, 156 weight parts of neopentyl glycol, and 0.34 weight part of tetrabutyl titanate were placed in a reactor equipped with a stirrer, thermometer, and an effluent cooler, and transesterification was performed for two hours at 170 to 220°C. As the temperature of the reaction system was raised from 220°C to 270°C after completion of transesterification, the pressure inside the system was gradually reduced to 500 Pa over 60 minutes. Polycondensation was then performed for 55 minutes at 130 Pa or below, and non-crystalline polyester (A) was obtained.

---

<sup>4</sup> Translator's note: The word "epoxy" reads "eboxy" in the original, which appears to be a typographical error.

[0059] According to NMR analysis of non-crystalline polyester (A), the dicarboxylic acid component was composed of 100 mol% of terephthalic acid, and the diol component was composed of 80 mol% of ethylene glycol and 20 mol% of neopentyl glycol. Also, the glass transition temperature thereof was 78°C, the number-average molecular weight was 28,000, and the acid value was 30 eq/10<sup>6</sup> g. Non-crystalline polyesters (B) through (E) were manufactured in the same manner as was non-crystalline polyester (A). The composition and results of measurement thereof are shown in Table 3. (Numerical values are mol% in resin.) Crystalline polyesters (a) through (e) were manufactured in the same manner as non-crystalline polyester (A). The composition and results of measurement thereof are shown in Table 3. (Numerical values are mol% in resin.)

[0060]

[Table 3]

		Synthesis Example									
		Non-crystalline polyester					Crystalline polyester				
		A	B	C	D	E	a	b	c	d	e
Acid	Terephthalic acid	100	90	88	90	100	63	51	68	65	68
	Isophthalic acid		10		10		10	25	11		32
	Adipic acid			12			27	24	21	35	
Glycol	Ethylene glycol	80	85	78	72	71				35	
	Diethylene glycol		15								
	Neopentyl glycol	20									
	2-methyl-1,3-propanediol			22							
	1,3-propanediol				28				86		
	Cyclohexane dimethanol					29					
	1,4-Butanediol						100	100		64	93
	1,6-Hexanediol								14		
	Polytetramethylene glycol (molecular weight: 1,000)									1	7
Properties	Number-average molecular weight	28000	32000	25000	33000	29000	18000	24000	30000	26000	34000
	Glass transition temperature (°C)	78	62	55	58	78	-6	-10	22	-9	-32
	Melting point (°C)	/	/	/	/	/	160	137	152	135	142
	Acid value (eq/10 <sup>6</sup> g)	30	25	16	38	25	28	32	28	30	32

[0061] The polyesters shown in Table 3 were mixed in a beaker with the components shown in Tables 4 and 5, and the mixture was kneaded on two 6-inch chilled rolls set to 180°C. The product was mixed while occasionally peeling off the resin adhering to the chilled rolls. When the resin did not peel off, it was assumed to have adhered to the rolls, and the time from the start

of mixing until adhesion was measured. After five minutes of kneading in the same manner, the roll interval was set to 0.3 mm (sheet thickness set to 0.3 mm), and a sheet was obtained. The results are shown in Tables 4 and 5.

[0062] Also, the lubricants and plasticizers indicated in Tables 4 and 5 refer to the compounds below.

I: tridecyl poly(oxyethylene)phosphate zinc chloride

II: styrene-modified polyethylene wax

III: tetrakis[methylene-3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]methane

Also, the numerical values for the quantities of polyesters, lubricants, and plasticizers in the tables represent weight parts.

[0063]

[Table 4]

		Working Example											
		1	2	3	4	5	6	7	8	9	10	11	12
Non-crystalline polyester	A	89	85	76.4								87	91.4
	B				93.9	83							
	C						79						
	D							87					
	E								92	84	87		
Crystalline polyester	a	10	14	22	5			5		15			
	b										12	12	
	c					16							
	d								7				
	e						20	7					7
Lubricant	I	1	1	1	1	1	1	1	1	1	1	1	1
	II			0.5									0.5
Stabilizer	III			0.1	0.1								0.1
Evaluation	Adhesion time (minutes)	18	22	>30	>30	28	27	24	11	10	13	15	>30
	Transparency	4	4	5	5	5	4	4	4	4	4	4	5
	Solvent resistance	5	5	5	4	4	5	5	4	4	4	5	4



[0064]

[Table 5]

		Comparative Examples			
		1	2	3	4
Non-crystalline polyester	A	90	99		
	B				
	C				
	D				
	E				
Crystalline polyester	a	10			52
	b			99	
	c				47
	d				
	e				
Lubricant	I		1	1	1
	II				
Stabilizer	III				
Evaluation	Adhesion time (minutes)	0	24	2	0
	Transparency	—	4	—	—
	Solvent resistance	—	1	—	—

[0065] The methods of evaluating transparency and solvent resistance in the tables were as follows.

Transparency: The sheets were compared visually and evaluated according to the following criteria. (5: extremely transparent; 4: good transparency; 3: transparent; 2: slightly opaque; 1: somewhat opaque.)

Solvent resistance: The presence of whitening and swelling were compared visually for sheets after ten minutes of dipping in methylethylketone, and evaluation was performed according to the following criteria. (5: no change; 4: some swelling, but no significant change; 3: whitening occurs; 2: whitening occurs and surface is slightly swollen; 1: whitening occurs and surface dissolves.)

In the section for evaluation of transparency and solvent resistance, "—" in the comparative examples indicates that a sheet could not be made due to adhesion.

[0066]

**[Effect of the Invention]** As described above, the polyester resin composition of the present invention consists of a polyester resin composition used for calendering comprising at least two types of polyester and a lubricant, and preferably consists of a composition in which a non-crystalline polyester, a crystalline polyester, and a lubricant are combined. Not only does using this composition make the heretofore difficult calendering of a polyester resin composition easy, but also it enables calendering of a high-quality sheet in which the finished sheet has good transparency and excellent solvent resistance and detergent resistance.

---

Continued from first page:

(51) Int. Cl. <sup>7</sup>	Class. Symbols	FI	Subject Codes (Reference)
B 29 L 7:00		B 29 L 7:00	
(72) Inventor: Tsuyoshi Hachitsuka			
c/o Toyobo General Research Institute			
1-1 Katata 2-chome, Otsu-shi, Shiga-ken			
F terms (reference):	4F071 AA44 AA84 AA86 AC15 AE11		
	BA01 BB04 BC01		
	4F204 AA24 AB07 FA07 FB02 FF01		
	4J002 CF031 CF032 EW046 FD176		
	GG02		